Final Report for Instrumentation for Measurement of Aerosol Differential Hygroscopic Growth Rates

Dean A. Hegg
University of Washington
Department of Atmospheric Sciences
Box 351640
Seattle, WA 98195-1640
phone: (206) 685-1984 fax: (206) 685-7160 email: deanhegg@atmos.washington.edu

David S. Covert
University of Washington
Department of Atmospheric Sciences
Box 354235,
Seattle, WA 98195-4235
phone: (206) 685-7461 fax: (206) 685-3397 email: dcovert@u.washington.edu

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LONG-TERM GOALS

This DURIP equipment grant is in support of our ongoing grant exploring the interactions of marine aerosols with clouds and the relationship between aerosol size and composition, and aerosol hygroscopicity and cloud condensation nuclei activity (Award # N00014-97-1-0132). Hence, these long-term goals of the "parent grant" are also those of this grant. However, the DURIP itself is more sharply focused on the fabrication of an instrument to address some of these long term goals, namely, obtaining reliable measurements of both super-micron aerosol particles and their hygroscopicity.

OBJECTIVES

Activity associated with this grant has concentrated primarily on the fabrication, testing, and deployment of our new aerosol hydration spectrometer (AHS). This instrument was deployed during the recently completed CARMA-III study and analysis of the data has just begun. While our specific objective for this grant has been the fabrication, testing and deployment of the AHS, we feel it important to list the two primary objectives for which the instrument has been used

- Refine measurements of super-micron aerosol in the MBL utilizing the AHS during the CARMA-III deployment.
- Explore aerosol hydration, and its relationship to composition for particles substantially larger than have previously been so characterized.

APPROACH

With the funding from the DURIP grant, we have designed and built an Aerosol Hydration Spectrometer. The spectrometer utilizes 90° white light scattering to avoid the size-response

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14. ABSTRACT
This DURIP equipment grant is in support of our ongoing grant exploring the interactions of marine aerosols with clouds and the relationship between aerosol size and composition, and aerosol hygroscopicity and cloud condensation nuclei activity (Award # N00014-97-1-0132). Hence, these long-term goals of the ?parent grant? are also those of this grant. However, the DURIP itself is more sharply focused on the fabrication of an instrument to address some of these long term goals, namely, obtaining reliable measurements of both super-micron aerosol particles and their hygroscopicity

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degeneracy problems associated with the coherent-light, forward scattering probes usually used to measure particles larger than a few microns in size. Two channels are employed, one operating at a high RH and one at close to ambient, to permit continuous measurement of the hygroscopic growth. The AHS can also be configured to explore the issue of delayed (or slow) condensational growth of particles to their equilibrium size. Some data on this issue have in fact been obtained but not yet analyzed. Of more immediate interest, size distribution data on particles out to ~ 9 μm diameter were obtained with the AHS from CARMA-III and have undergone preliminary analysis. As part of this task, we found it necessary to better characterized the aerosol inlet used on the Twin Otter research aircraft and did so in a major laboratory stuff conducted prior to the CARMA-III deployment. Results of this study are also reported below.

While the methodology for refined measurement of the super-micron aerosol size distribution (objective two) is implicit in the design of the AHS itself, measurement of the hygroscopicity of this aerosol and its relationship to chemical composition is more involved. For hygroscopicity, the main issue is the unambiguous association of the dry and wet diameters from two separate size distribution measurements. We have adopted an approach developed at the Institute for Tropospheric Research in Leipzig. Called the Descriptive Hygroscopic Growth Factor (DHGF) approach, it does permit recovery of either the wet or dry distribution from measurement of the other. For the association of the measured hygroscopicity as a function of size with chemical composition, we use several different filter-based approaches for measuring the aerosol chemistry. Most novel is the use of the Pacific Northwest National Laboratory (PNNL) TRAC sampler. This device is essentially an automated SEM/TEM substrate sampler that can expose substrates as frequently as every 30 s. The substrates are analyzed post-flight by a variety of powerful analytical techniques (cf., Laskin et al, 2003; Hoffman et al, 2004). These techniques yield size-resolved chemical composition data to collate with the size-resolved hygroscopicity data from the AHS.

WORK COMPLETED

The AHS has been fabricated, tested and successfully deployed in CARMA-III. Size distribution measurements obtained with it address the second of our goals though clearly more data would be desirable. Data have also been obtained from CARMA-III on the size resolved hygroscopicity of marine aerosol and its relationship to composition. Analysis will take place over the next year.

RESULTS

The first result of the activity undertaken with this grant worthy of note is the determination of the transmission efficiency of the Twin Otter aerosol inlet. This finding is very important because few aerosol inlets in atmospheric research are well characterized for supermicron particles and it is essential that we establish that the one delivering particles to the AHS in fact passes larger particles. The measured transmission efficiency is shown in Figure 1.

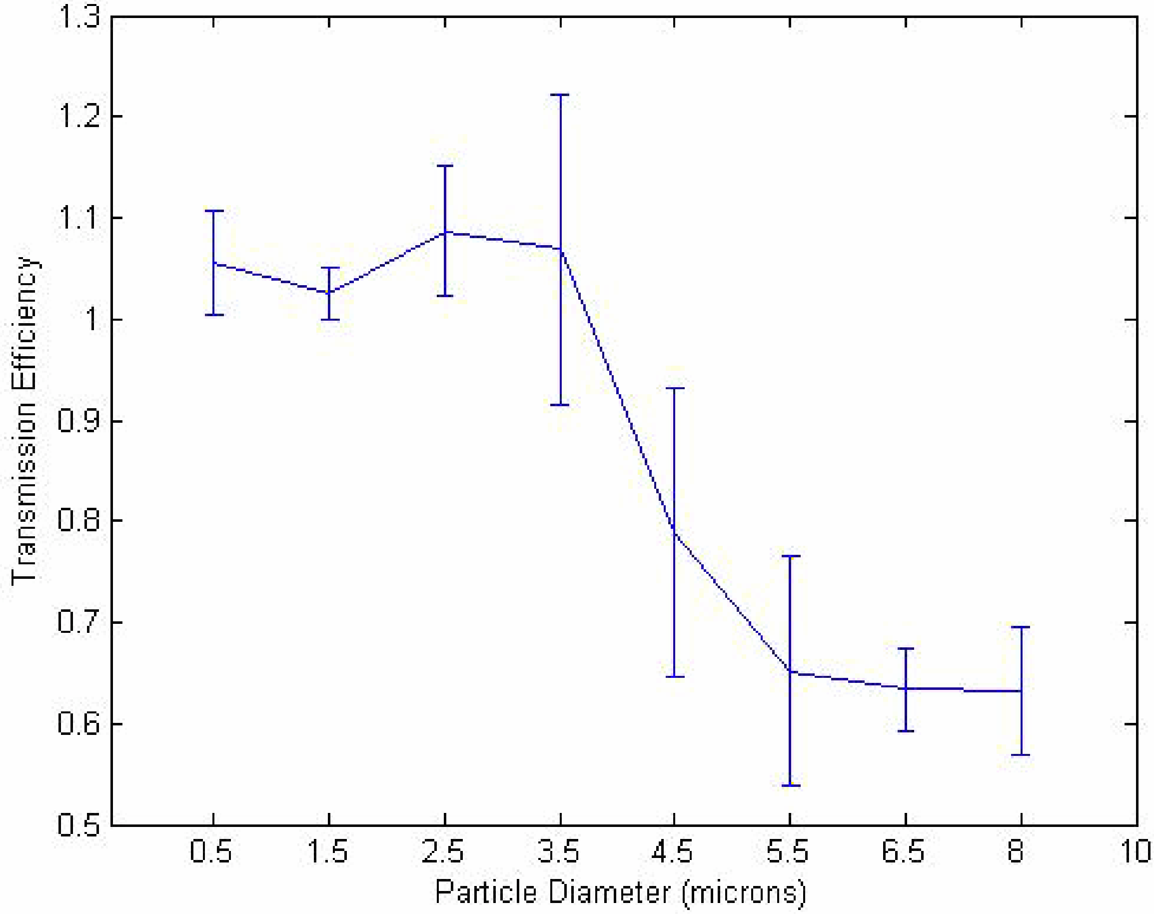

Figure 1. The mean transmission efficiency for particles from 0.5 to 9 microns in diameter. The sigmoid curve is typical of aerosol inlets though the plateau above ~ 5 μm diameter is higher than expected (60% efficiency) and indicates sub-isokinetic flow.

The essentially 100% efficiency up to ~ 3.5 μm diameter is excellent and the plateau above 5 μm is higher than expected, possibly indicating sub-isokinetic flow. This characterization provides the basis for accurate recovery of super-micron particle size distributions in the MBL.

Utilizing the above inlet together with the AHS, particle size distributions were recovered from the CARMA-III data set. An example of such distributions is shown in Figure 2.

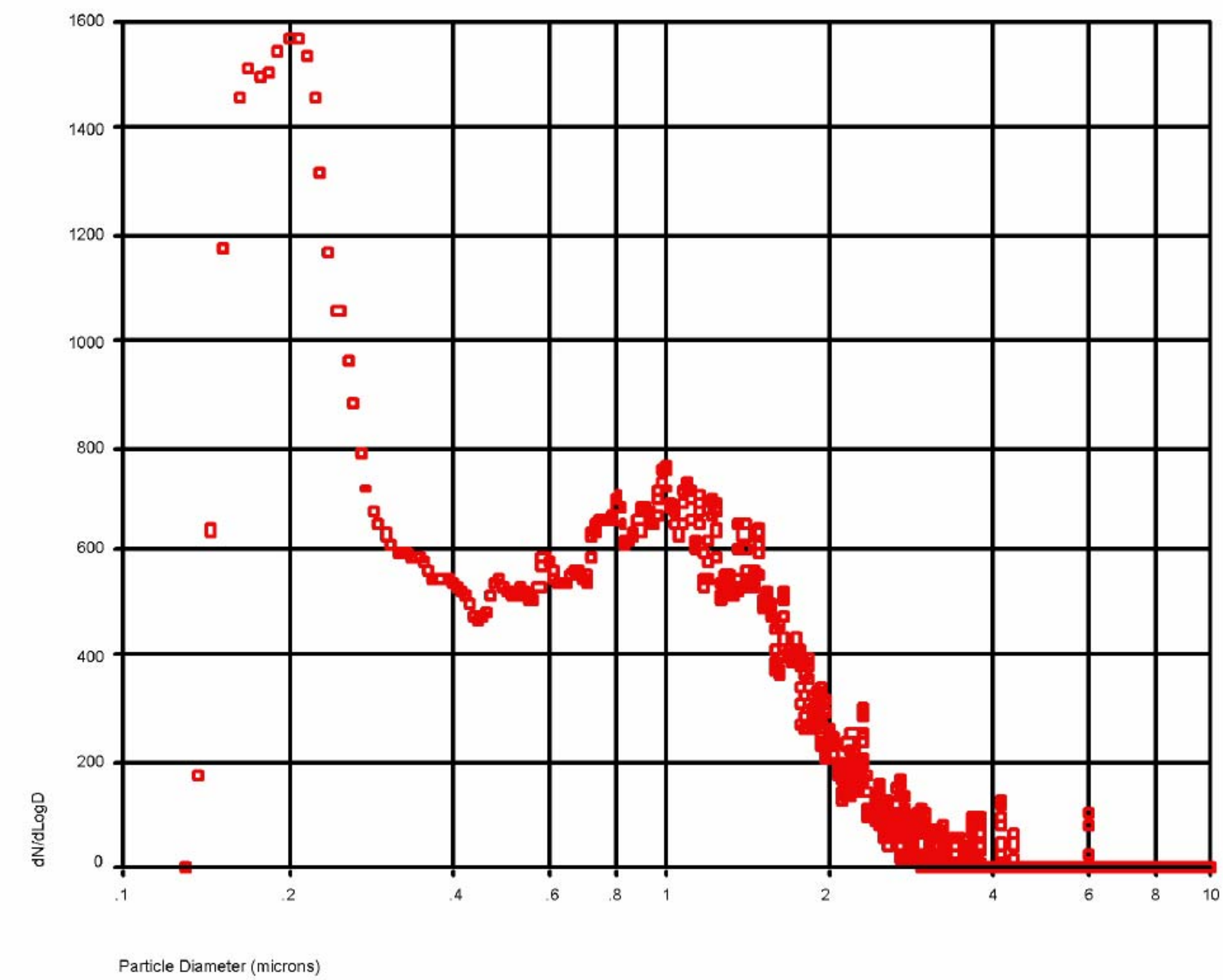

Figure 2. The dry size distribution measured by the AHS at 30 m AMSL during the flight of August 23rd. The wind speed was ~ 10 m s^{-1}. The particle data below 0.25 μm is suspect and thus the smaller size mode centered at 0.2 μm is should not be taken too seriously. However, the mode at 1.0 μm is quite real.

The smaller size mode at 0.2 μm is somewhat suspect since the AHS data are currently questionable below about 0.25 μm. However, the mode at 1.0 μm is quite real and leads to a volume mode at ~ 2 μm. This is of some significance since chemical analysis for this date suggests that most of the particulate mass is sea salt. Both the concurrent FSSP data from this study and many previous measurements show a FSSP volume mode for sea salt in the 5-6 μm range, in contrast to APS and impactor data which find the mode in the 1-3 μm range. The normal FSSP mode is considered an artifact of the FSSP (cf. Reid et al, 2005) and the AHS seems to avoid this problem, as we had hoped.

In addition to providing sizing data, the AHS permits the measurement of the aerosol hygroscopicity as a function of size out to larger sizes than have previously been investigated. Examples of the aerosol hygroscopicity for sea salt measured on three different dates are shown in Figure 3.

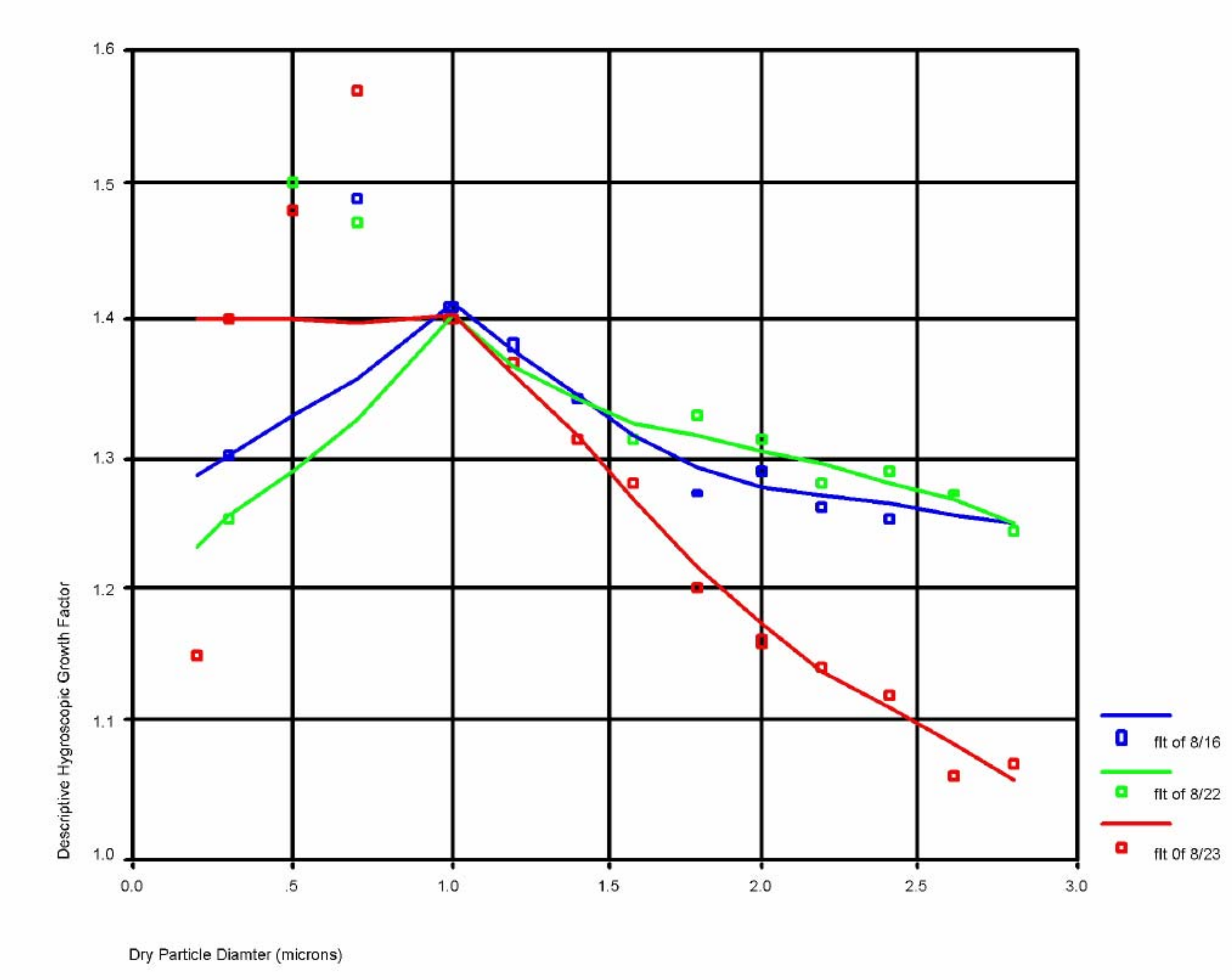

Figure 3. Descriptive Hygroscopic Growth Factors for three different sea salt samples. The lines associated with the data points are Lowess fits. The rise in the magnitude of the DHGF from 0.2 to 1 μm is similar to previous observations but the fall off above 1 μm is unexpected, particularly the sample of 8/23.

For particle sizes below roughly 0.5 μm, the growth factors are in reasonable accord with numerous previous studies (e.g., Berg et al, 1998), including the slight increase in the growth factor with increasing size. However, the peak at ~ 1 μm and the subsequent monotonic decrease in growth factor with increasing size are hitherto unknown phenomena. In part, the decline is associated with changes in the index of refraction as the particles likely have a larger water component but this is probably not the whole story. The contrast between the case of 8/23 and the other two cases is also noteworthy. The lower hygroscopicity of the 8/23 sample appears to be explicable in terms of a differing source area. HYSPLIT back trajectories reveal that, unlike the two other cases that have purely marine back trajectories, the 8/23 trajectory passes over southern Oregon, an area experiencing wild fires during our observational period. The presence of pyrogenic aerosols could explain the reduced hygroscopicity. Indeed, a sample of a fire aerosol plume from the same region, sampled some days earlier,

demonstrates this and is shown in Figure 4. In any case, these preliminary findings are quite exciting and call for further analysis.

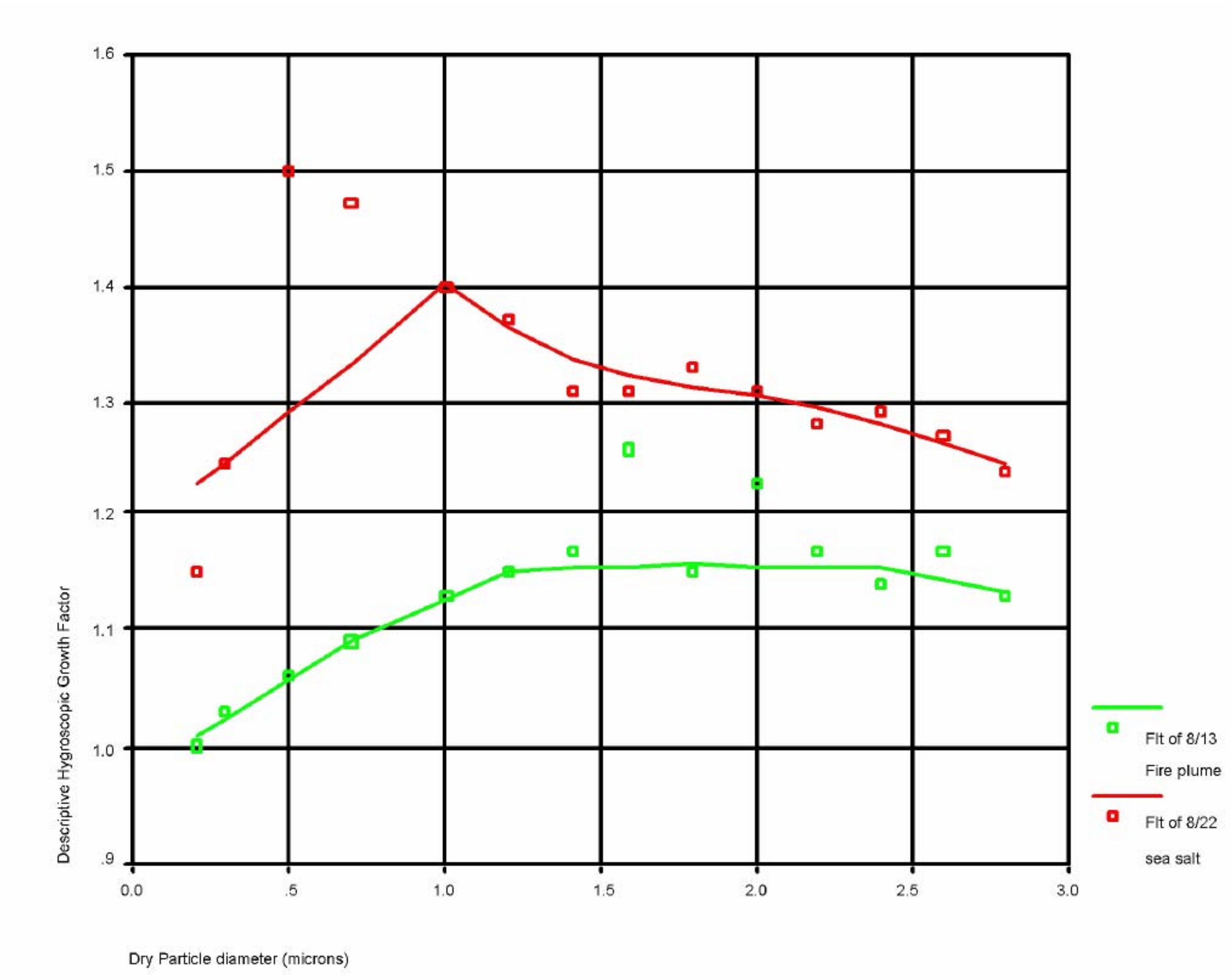

Figure 4. DHGF's for the fire plume encountered on 8/13. Note the much lower values for the hygroscopic growth compared to the sea salt case of 8/22.

IMPACT/APPLICATIONS

These results demonstrate that the AHS is functioning properly and provides a useful tool for exploring aerosol hygroscopicity. The hygroscopicity data as a function of aerosol size reveal that there are distinct patterns of DHGF for different aerosol type and illustrate the importance of chemical composition in the determination of aerosol hygroscopicity. The first measurements of size dependent hygroscopicity for super-micron particles are intriguing and detailed analysis should prove very interesting.

TRANSITIONS

None.

RELATED PROJECTS

The size dependent hygroscopic growth of aerosols, including super-micron aerosols (cf., Quinn et al, 1998), is a major factor in both the radiative energy balance of the lower marine atmosphere and the propagation of radiation through the MBL. Such radiation properties are necessary parameters for numerical modelers developing prognostic models. It is, furthermore, an aerosol characteristic closely related to CCN activity and, indeed, such activity can be predicted from it. Hence, these measurements are highly relevant to determination of CCN spectra and thus of the microphysics of MBL clouds.

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PUBLICATIONS

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PATENTS

None.